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C3W W121 W122
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(56) Documents cited
GB 2163348 A EP 0374658 A2 EP 0177158 A2
JP 630221102 A

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(54) **Adhesive**

(57) An adhesive composition suitable for use as a wallpaper adhesive comprises a mixture of a water-soluble adhesive polymer such as a cellulose derivative or starch and a product (such as a composite of a dry acid and a carbonate or bicarbonate) reactive in the presence of water to release a gas. The mixture which is conveniently in compressed tablet form has a moisture content sufficiently low to prevent significant reaction in the product before immersion of the mixture in water. The dry acid may be citric, sulphamic or tartaric acid.

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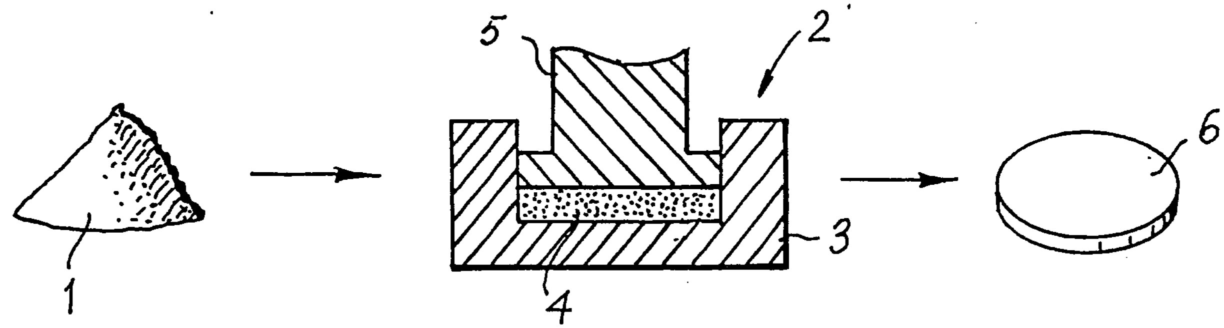

FIG.1

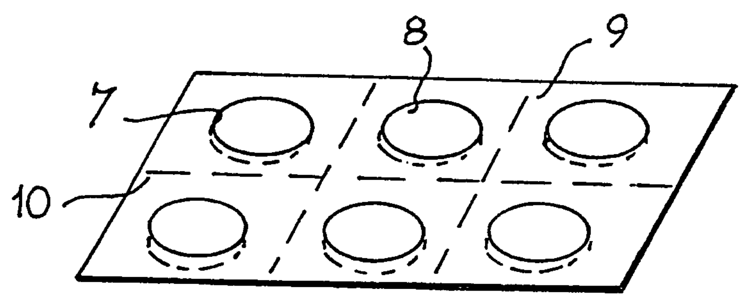

FIG.2

2/2

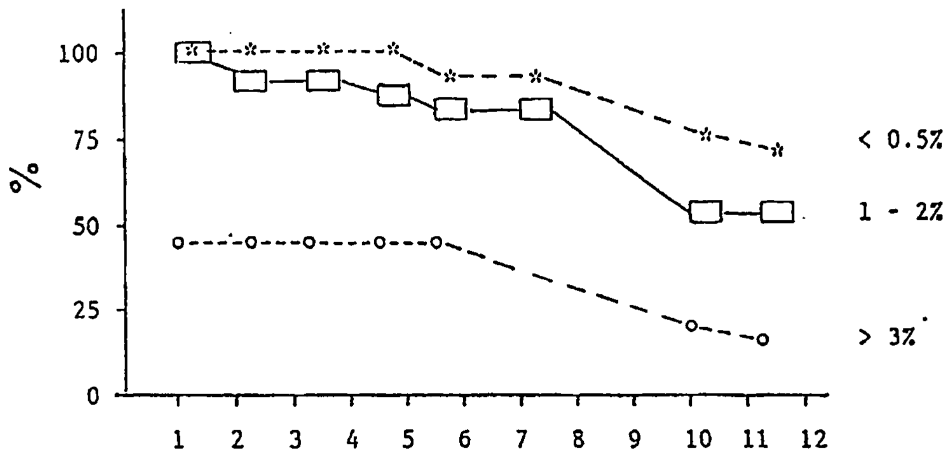
100
75
50
25
0
%
1 2 3 4 5 6 7 8 9 10 11 12
< 0.5%
1 - 2%
> 3%

FIG. 3

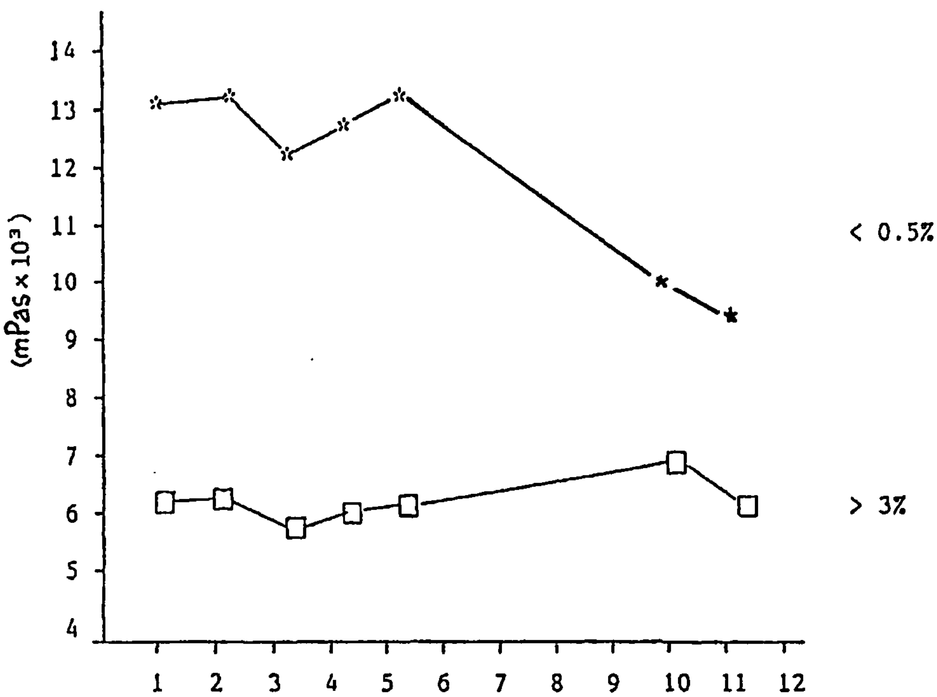
14
13
12
11
10
9
8
7
6
5
4
(mPas × 10³)
1 2 3 4 5 6 7 8 9 10 11 12
< 0.5%
> 3%

FIG. 4

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Adhesive

This invention relates to adhesives and has particular reference to adhesive compositions based on water-soluble adhesive polymers. It has further particular reference to water-based adhesive polymers for use as wallpaper adhesives.

Water-soluble adhesives for use as wallpaper adhesives are well known. Originally such adhesives were based on starch but in more recent years cellulosic-based adhesive polymers have been developed and have been widely used. These cellulosic materials, typically cellulose ethers, form very good wallpaper pastes but there are problems in obtaining a solution of the cellulose ether in the water to form the paste. If attempts are made to dissolve large particles of cellulose ethers they are slow to dissolve. If on the other hand the cellulose ethers are ground into very fine particles, the particles tend to agglomerate and form lumps which again are slow to dissolve.

The most normal solution to this problem is to partially cross-link the cellulose ether powders and to mix the cellulose ether powders with a product which will raise the pH of the water to break down the cross-linkages. Normally, sodium carbonate or sodium bicarbonate is used as the pH-raising material.

The presence of the partial cross-linking avoids the formation of instant lumps when dissolving commences and the sodium carbonate or sodium bicarbonate raises the pH to gradually break down the partial cross-linking to enable the cellulose ether to pass into solution.

Proposals have also been made to boost the viscosity of the wallpaper paste by incorporating superabsorbers which are typically more heavily cross-linked cellulose ether materials which absorb significant quantities of the available water leaving only the interstitial water to be

thickened by the dissolving polymer. In effect with this type of adhesive the concentration of the dissolving polymer is greater and hence the apparent viscosity is higher. Proposals have been made to make more rapidly dissolving wallpaper pastes and so called "instant" pastes have been developed. These pastes are not truly instant in that they are only ready for use within three to fifteen minutes from mixing.

Further proposals have been made, see DE-A-3,103,338, to produce quickly soluble pastes by adding water to very fine, virtually dust-like, water-soluble cellulose ethers in a granulator to increase the grain size of the particles. Typically, 40% moisture content is added to the very fine particles and these particles are granulated, followed by drying to a residual moisture content of typically 10%. The granulator-produced cellulose ethers containing a moisture content of 10% may be processed into a tablet using a tablet press.

By the present invention there is provided an adhesive composition comprising a dry mixture of a water-soluble adhesive polymer and a product reactive in the presence of water to release a gas.

By the term dry as used herein is meant having a moisture content sufficiently low to prevent significant reaction in the product before immersion of the mixture in water.

Preferably, the moisture content of the mixture is less than 1%, or less than 0.75% or less than 0.5%, even more preferably the moisture content is less than 0.25% or less than 0.2% or less than 0.15% or less than 0.1% or less than 0.05%. All percentages used herein are weight percentages.

The product reactive in the presence of water may be formed of two components and may comprise a dry acid component and a carbonate or bicarbonate component. The

acid may be selected from citric acid, sulphamic acid and tartaric acid. The carbonate or bicarbonate is preferably sodium carbonate. Alternatively the carbonate or bicarbonate may be a salt of potassium, lithium or ammonium.

The adhesive polymer may be a cellulose derivative, a starch, a modified starch or a mixture of two or more thereof. The cellulose derivative may be a cellulose ether, preferably methyl cellulose, carboxymethyl cellulose, hydroxyethylmethyl cellulose, ethylhydroxyethyl cellulose or hydroxypropylmethyl cellulose. The cellulose derivative may be lightly cross-linked so as to be soluble at a pH greater than 8 and to delay dissolution in water. The cross-linking may be effected by glyoxal additions. There may be an excess of the basic component such as the carbonate over the acid component to provide a pH not less than 8 in the solution after dissolving. Preferably, the pH is in the range 8 to 9.5.

Some of the cellulose ether material may be crosslinked so as to be water-insoluble and form a superabsorber. Some of the cellulose ether may be partially crosslinked to delay the onset of solution during immersion in water. Some or all of the cellulose ether may be in the form of granules or powder.

The adhesive composition may be compressed into a tablet. In the case of a tablet the total moisture content of the mixture is preferably less than 0.25%. Optimally the pressing load for the tablet should not exceed about 1000 kg/cm^2. The tablet should be pressed in dry atmosphere and is preferably sealed in a moisture-impermeable container or wrapping immediately after pressing. A plurality of tablets may be incorporated into a single container or the container may be in the form of a multi-cell blister pack, each cell of the pack containing a single tablet. The water-soluble adhesive may comprise a mixture of cellulose- and starch- based adhesives.

The tablets may be such a size that, when mixed with a volume of water selected from the group 200 ml, 250 ml, 300 ml, 330 ml, 350 ml, 500 ml, 1 litre, 2 litres and 5 litres or ½ pint (0.3 litre), 1 pint (0.6 litre), 2 pints (1.1 litres), 4 pints (2.3 litres) or 8 pints (4.5 litres), an individual tablet produces an acceptable quality of wallpaper adhesive.

The tablet may be cubic or a parallelepiped or may be in the form of a disc. The disc may be circular or oval in cross-section. The tablet may incorporate break-up aids such as a starch, a superabsorber, silica or talc. The tablet may incorporate one or more binding agents such as a light mineral oil or starch.

By way of example, embodiments of the present invention will now be described with reference to the accompanying drawings, of which:-

Figure 1 is a schematic view of a tableting procedure,

Figure 2 is a perspective view of a multi-component blister pack,

Figure 3 is a graph of performance against shelf life, and

Figure 4 is a graph of viscosity against shelf life.

Referring to Figure 1, this shows a pile of dry mixture 1 which is inserted into a press generally indicated by 2. The press incorporates a die component 3 into which the powder is located as at 4 and a ram 5 which is pressed onto the powder 4 to form a tablet such as tablet 6. Immediately after manufacture the tablets are transferred to a series of blister pack receptacles 7,8 etc. and the blister pack is then sealed with a cover layer. Typically, the blister pack would be made of a water-impermeable plastics material and there may be provided tear lines such as dotted lines 9,10 to permit an individual tablet to be

removed from the blister pack.

In a first series of tests four formulations were mixed in the form of powders. Details of the mixtures are given in Table 1 below.

TABLE 1

Formulation	Pressure	Tablet Strength	Dissolution Time	Thickening Characteristics
1) B2/15-(67%) Na_2CO_3 + $NaHCO_3$ + C/A (28%) A250-(5%)	N.A.	N.A.	1 min	2 - 3 mins Entrapped air bubbles give Turbid Appearance (Viscosity 4500 mPas)
2) B2/15-(69%) Na_2CO_3 + $NaHCO_3$ + C/A (31%)	N.A.	N.A.	1 min	4 - 5 mins Entrapped air bubbles
3) HPM 30000-(67% Na_2CO_3 + $NaHCO_3$ + C/A (28%) A250-(5%)	N.A.	N.A.	1 min	4 - 5 mins Grainy Appearance (Viscosity 11,300 mPas)
4) HPM 30000-(69%) Na_2CO_3 + $NaHCO_3$ + C/A (31%)	N.A.	N.A.	1 min	4 - 5 mins Grainy Appearance (Viscosity 12,000 mPas)

Formulation	Pressure	Tablet Strength	Dissolutio Time	Thickening Characteristics
2) B2/15-(69%) Na_2CO_3 + $NaHCO_3$+ (31%) C/A	3 MT	Weak Edges	-	-
3) HPM 30000-(67%) Na_2CO_3 + $NaHCO_3$+ (28%) C/A A250-(5%)	3 MT	Weak Edges	-	-
4) HPM 30000-(69%) Na_2CO_3 + $NaHCO_3$+ (31%) C/A	3 MT	Weak Edges	-	-

In Table 1 B2/15 is a reference to Celacol HPM 15000 P (100 mesh) - which is hydroxypropylmethyl cellulose of less than 150 micron particle size with a 2% viscosity in water of 15000 milli Pascal second (mPas) when measured using an Ostwald viscometer thermostatted at 20°C. C/A refers to citric acid and the ratio of sodium carbonate to sodium bicarbonate to citric acid was in all cases 1 part sodium carbonate to 7 parts citric acid to 11 parts sodium bicarbonate. A250 refers to a fine-powder carboxymethyl-cellulose superabsorber with a capacity to absorb about 50 times its own weight of de-ionised water. HPM 30,000 identifies hydroxypropylmethyl cellulose of such a molecular weight that it forms a viscosity of between 1300 and 1700 mPas at a 1% solution in deionised water when measured using an Ostwald viscometer thermostatted at 20°C. N.A. means not applicable.

From Table 1 it can be seen that all four formulations dissolved extremely rapidly and thickened within 2 to 5 minutes. When the powder was poured into the water at a standard rate of 2.98 g powder to 100 cc water the powder fizzed and dissolved very quickly as can be seen from Table 1.

No attempt was made in connection with the powder formulations of Table 1 to control the water content of the powders. Attempts were then made to formulate tablets from the formulations 2,3 and 4 referred to above. 8 g of powder was placed into a tableting press and a pressure of 3 metric tons (3 MT) was exerted onto the powder to form a tablet. As can be seen from Table 1, tablets were produced but they had weak edges. When these tablets were added to water they started to dissolve but formed a lumpy mixture with a gel-like exterior which prevented full dissolution. The tablets were therefore working partially but were not sufficiently effective for these formulations to be used in tablet form.

As a result of the inconsistencies discovered in connection with the work carried out to produce the information identified in Table 1 a further series of tests were carried out in which all of the powdered ingredients were dried for 24 hours at 40°C and then stored over a silica desiccant. It was also decided to omit the sodium bicarbonate component, using only sodium carbonate, as sodium bicarbonate tends to be more moist than sodium carbonate. Anhydrous citric acid was used for the starting material component.

The formulations used are set out in Table 2 and reference in that Table to Celacol WA is a reference to a hydroxypropylmethyl cellulose powder lightly cross-linked with glyoxal and having a 2% viscosity (when dissolved in water) of between 11500 and 16000 mPas measured using an Ostwald viscometer thermostatted at 20°C. B.U. refers to a hydroxypropylmethyl cellulose of extremely fine particle size and having a 2% viscosity (when dissolved in water) of

between 68,000 and 92000 mPas, measured using an Ubbelohde viscometer thermostatted to 20°C, and C/A means citric acid. The tablets were manufactured this time using 5 metric tons loading on the press.

The result of maintaining very low moisture contents was dramatic improvement in the dissolution characteristic of the tablets, as can be seen in Table 2 below, even after the tablets had been stored for several days.

Table 2

FORMULATION	PRESSURE	TABLET STRENGTH	DISSOLUTION	THICKENING CHARACTERISTICS
CELACOL WA (48%) C/A (Anhydrous) (20%) Na_2CO_3 (32%)	5 MT	Poor Binding	1-2 min (30 sec. stirring)	pH = 9.2 Viscosity (cps): 5 min 9,000; 7 min 11,000; 8 min 13,100 Smooth solution. No bubbles or lumps retained.
B2/15 (48%) C/A Anhydrous (20%) Na_2CO_3 (32%)	5 MT	Poor Binding	3-4 min (30 sec. stirring)	pH = 9.0 Viscosity (cps): 5 min 3,200; 7 min 4,000; 8 min 4,600 Small lumps remain, which disappear after 30 minutes.
B.U. (47.95%) C/A Anhydrous (20%) Na_2CO_3 (32%) Light Mineral Oil (0.05%)	5 MT	Poor Binding	2-3 min (30 sec. stirring)	Viscosity (cps): 5 min 8,400; 7 min 9,000; 8 min 11,600 pH = 9.2
Celacol WA (47.95%) C/A Anhydrous (20%) Na_2CO_3 (32%) Light Mineral Oil (0.05%)	5 MT	Slightly improved Binding	2-3 min (30 sec. stirring)	Viscosity (cps): 5 min 8,500; 7 min 9,300; 8 min 12,600 pH = 9.4

All powdered ingredients dried for > 24 hours at 40°C.

It can also be seen that the viscosity, with the exception of the B2/15 formulation, was very acceptable after thickening had occurred. With the realisation that significant effects on the tableting characteristics of the mixture were to be obtained by varying the moisture content, a further series of tests was carried out.

To investigate the effect of moisture content on the tableted formulations, a series of tablets was produced from a starting material comprising 45 g Celacol WA, 18 g anhydrous citric acid, 30 g sodium carbonate and 0.1 g light mineral oil. All the powdered ingredients had been dried for 24 hours at 40°C and stored over silica desiccant. The tablets were then divided into three batches. The first batch comprising 16 tablets, was immediately sealed in individual heat-sealed polyethylene film bags. The second batch, again comprising 16 tablets, was allowed to equilibriate at room temperature until a moisture content in the range 1 to 2% was reached and again these were then heat sealed in individual polyethylene film bags. The third batch comprising 15 tablets was permitted to equilibriate at room temperature to reach a moisture content in excess of 3% and these tablets were then also heat sealed in individual polyethylene film bags.

All tablets were left untouched and some of each batch were tested for performance at intervals of about 1 week for 6 or 7 weeks, two tablets being tested after having been left on the shelf in the laboratory for 10 and 11 weeks respectively.

For testing purposes each tablet was removed from its bag by cutting open the bag and extracting the tablet with tweezers. The tablet was then dropped into deionised water at 20°C so that the concentration of the adhesive component of the tablet in water would be 2%. Tablet performance was assessed by estimating the percentage of tablet remaining undissolved after 2 minutes. Successful tablets would normally break up and dissolve in under 1 minute.

normally break up and dissolve in under 1 minute.

Figure 3 is a graph of performance against shelf life for the three batches of the tablets. It can be seen that the performance of the tablets having an initial moisture content of less than 0.5% is excellent for 5 weeks but then falls slowly to about 75% after 12 weeks. The performance of tablets having an initial moisture content of 1 to 2% falls from the 1 week level fairly continuously for 7 weeks and then fairly dramatically so as to plateau at something between 50 and 55% performance after 11 to 12 weeks.

It can be seen, by comparison, that tablets having an initial moisture content in excess of 3% have a poor performance even after 1 week in the heat-sealed bag and this performance deteriorates further, so that after 10 to 11 weeks the performance is less than 25%.

Measurements were also made of the viscosity of the solution 8 minutes after immersion of the tablet in water. A Brookfield Viscosimeter was used and the results of the tests on the tablets having an initial moisture content of less the 0.5% and an initial moisture content of greater than 3% are shown in Figure 4. These show that the viscosity of the solutions manufactured from tablets having an initial moisture content of less than 0.5% is normally of the order of 13,000 mPas but falls to approximately 11,000 mPas after 11 weeks. By comparison the viscosity of the solutions produced from tablets having an initial moisture content of greater than 3% never varies very much and is always in the range 5500 to 7,000 mPas.

It was observed that the reduction in performance of the tablets was associated with failure of the tablets to break-up and the development of a mucous film around the partially broken up tablets which prevented further dissolution of the tablets or of the broken-off portions of the tablets.

Tables 3 and 4 below show the viscosities developed after 8 minutes using the tablets having an initial moisture content of less than 0.5% (Table 3) and having an initial moisture content of greater than 3% (Table 4). The information contained in Tables 3 and 4 has been used to generate Figure 4.

TABLE 3

MOISTURE CONTENT <0.5%

WEEKS	VISCOSITY mPas
1	13,000
2	13,200
3	12,100
4	12,400
5	13,000
10	10,000
11	9,300

TABLE 4

MOISTURE CONTENT >3%

WEEKS	VISCOSITY mPas
1	6,050
2	6,100
3	5,700
4	5,900
5	6,200
10	6,500
11	6,100

To further investigate the moisture contents of the tablets the exact moisture levels of the remaining tablets of the three batches of tablets were measured. Moisture contents were measured initially from a scraping from each tablet before it was sealed in the polyethylene bag, and finally for tablets just after removal from the heat- sealed polyethylene bags. Set out in Table 5 below are the results for the remaining tablets from each of the three batches showing the initial moisture contents (I) and the final moisture contents (F) after the tablets had been permitted to remain in the heat sealed bags for the time indicated in weeks in the left hand column of Table 5.

TABLE 5

MOISTURE CONTENTS

	BATCH 1		BATCH 2		BATCH 3	
	Moisture Content < 0.5%		Moisture Content 1-2%		Moisture Content > 3%	
WEEKS	I	F	I	F	I	F
1	0.08	0.08	1.13	1.13	3.39	3.39
2	0.09	0.09	1.20	1.29	3.31	3.72
3	0.22	0.21	1.35	1.37	3.10	3.15
4	0.11	0.24	1.15	1.19	3.41	4.10
5	0.08	0.38	1.39	1.87	3.45	4.19
6	0.16	0.37	1.57	2.14	3.31	4.25
7	---	---	---	---	---	---
8	---	---	---	---	---	---
9	---	---	---	---	---	---
10	0.13	1.11	1.29	2.25	3.25	3.95
11	0.14	1.14	1.35	3.39	3.71	4.51

It can be seen that there is a gradual increase in moisture content, due to permeation of moisture through the walls of the bag as a result of diffusion. It can further be seen that the moisture content for the very dry tablets increases more as a percentage than for the more moist tablets. The results of the performance assessed as percentage of tablet dissolved after 2 minutes against periods are set out in Tables 6, 7 and 8 below.

TABLE 6

Moisture Content < 0.5%

WEEK	PERFORMANCE
1	100
2	100
3	100
4	100
5	95
6	95
10	75
11	70

TABLE 7

Moisture Content 1 - 2%

WEEK	PERFORMANCE
1	100
2	95
3	95
4	90
5	80
6	80
10	50
11	50

TABLE 8

Moisture Content > 3%

WEEK	PERFORMANCE
1	45
2	45
3	40
4	40
5	40
6	--
10	25
11	20

It can be seen that the performance of the initially very dry tablets (Table 6) starts to fall off after four weeks, which corresponds to a moisture content of about 0.24%. From this information it has been determined that a preferred maximum moisture content for the tablets of this particular formulation should not exceed 0.25%.

Because the components added to the formulation to produce the gas may affect the final viscosity of the solution it may be desirable to use mixtures of a cellulose derivative and starch. In a further test, 6 g of granular carboxymethyl cellulose having a molecular weight such that it gave a viscosity of 2,000 millipascal seconds at 1% solution in deionised water measured using an Ostwald viscometer thermostatted at 25°C was mixed with 2 g cold-water maize starch plus 0.5 g anhydrous citric acid and 0.75 g sodium carbonate. This gave a good disintegrating tablet which had a high viscosity in the eventual solution.

CLAIMS

1. An adhesive composition characterised in that it comprises a dry (as herein defined) mixture of a water-soluble adhesive polymer and a product reactive in the presence of water to release a gas.

2. A composition as claimed in claim 1 further characterised in that contains an amount of water of less than 0.05% or less than 0.1% or less than 0.15% or less than 0.2% or less than 0.25% or less than 0.5% or less than 1% by weight.

3. A composition as claimed in claim 1 or 2 further characterised in that the product reactive in the presence of water is formed of two components.

4. A composition as claimed in claim 3 further characterised in that one of the two components is a dry acid component and the other component is a carbonate or bicarbonate.

5. A composition as claimed in claim 4 further characterised in that the dry acid is selected from citric acid, sulphamic acid and tartaric acid.

6. A composition as claimed in claim 4 or 5 further characterised in that the carbonate or bicarbonate is a salt of an alkaline material selected from the group sodium, potassium, lithium or ammonium.

7. A composition as claimed in any one of claims 1 to 6 further characterised in that the adhesive polymer is selected from:-

(i) cellulose derivatives, preferably a cellulose ether, further preferably selected from:

a) methyl cellulose

b) carboxymethyl cellulose

c) hydroxyethylmethyl cellulose
d) ethylhydroxyethyl cellulose, and
e) hydroxypropylmethyl cellulose,

(ii) starch
(iii) modified starches, and
(iv) mixtures of two or more of such adhesive compositions.

8. A composition as claimed in claim 7 further characterised in that it contains a cellulose ether which is lightly cross-linked so as to be soluble at a pH not less than 8, preferably in the range 8 to 9.5.

9. A composition as claimed in claim 8 further characterised in that there is an excess of the carbonate or bicarbonate component to provide the pH not less than 8 in the solution after dissolving.

10. A composition as claimed in any one of claims 7 to 9 further characterised in that it contains a cellulose ether, some of which is cross-linked so as to be water-insoluble to form a super-absorber.

11. A tablet characterised in that it is a dry compressed composition as claimed in any one of claims 1 to 10.

12. A tablet as claimed in claim 11 further characterised in that it is compressed at a load not exceeding 1000 kg/cm².

13. A tablet as claimed in claim 11 or 12 further characterised in that it is of such a size that, when mixed with a volume of water selected from 200ml, 250ml, 300ml, 330ml, 500ml, 1 litre, 2 litres and 5 litres or ½ pint (0.3 litre), 1 pint (0.6 litre), 2 pints (1.1 litres), 4 pints (2.3 litres) and 8 pints (4.5 litres), it produces an acceptable quality of wallpaper adhesive.

14. A tablet as claimed in any one of claims 11 to 13 further characterised in that it includes one or more binding agents, particularly a light mineral oil.

15. A tablet as claimed in any one of claims 11 to 14 further characterised in that it includes silica or talc.

16. A container of a plurality of tablets characterised in that they are as claimed in any one of claims 11 to 15.

17. A container in the form of a multi-cell blister pack, each cell of the pack containing a single tablet characterised in that each tablet is as claimed in any one of claims 11 to 15.

18. Wallpaper adhesive, optionally in tablet form, substantially as herein defended with reference to and as illistrated by the accompanying drawings.

ZC

Patents Act 1977
Examiner's report to the Comptroller under Section 17 (The Search Report) CORRECTED

Application number 9200116.3

Relevant Technical fields

(i) UK Cl (Edition K) C3V (VAL,VCX,VDX,VEX); C3A C3L (LDT,LDJ,LDK)

(ii) Int Cl (Edition 5) C08K; C09J

Search Examiner: B J BALDOCK

Databases (see over)

(i) UK Patent Office

(ii) ONLINE DATABASES : DERWENT WPI, WPIL

Date of Search: 19 FEBRUARY 1992

Documents considered relevant following a search in respect of claims 1 TO 18

Category (see over)	Identity of document and relevant passages	Relevant to claim(s)
X	EP037465 A2 WOLFF WALSRODE	1,3,4,5
X	EP 0177158 A2 B P CHEMICALS see Claims 1 to 3, page 3 lines 1-15	1,5,7
X	GB 2163348 A DENTAB	1,3 to 7
X	* JP 630221102 A SANWA SHOJI see WPI Accession No: 88-302684/43	1,3,4, 5,7

* Indicates corrected entry

SF2(p)

tp - c:\wp51\doc99\fil000234

Category	Identity of document and relevant passages	Relevant to claim(s)

Categories of documents

X: Document indicating lack of novelty or of inventive step.

Y: Document indicating lack of inventive step if combined with one or more other documents of the same category.

A: Document indicating technological background and/or state of the art.

P: Document published on or after the declared priority date but before the filing date of the present application.

E: Patent document published on or after, but with priority date earlier than, the filing date of the present application.

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